Macroscopic quantum coherence in antiferromagnetic molecular magnets

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The macroscopic quantum coherence in a biaxial antiferromagnetic molecular magnet in the presence of magnetic field acting parallel to its hard anisotropy axis is studied within the two-sublattice model. On the basis of instanton technique in the spin-coherent-state path-integral representation, both the rigorous Wentzel-Kramers-Brillouin exponent and preexponential factor for the ground-state tunnel splitting are obtained. We find that the quantum fluctuations around the classical paths can not only induce a new quantum phase previously reported by Chiolero and Loss (Phys. Rev. Lett. 80, 169 (1998)) , but also have great influnence on the intensity of the ground-state tunnel splitting. Those features clearly have no analogue in the ferromagnetic molecular magnets. We suggest that they may be the universal behaviors in all antiferromagnetic molecular magnets. The analytical results are complemented by exact diagonalization calculation.

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I. INTRODUCTION

In recent years, owing mainly to the rapid advances both in new technologies of miniaturization and in highly sensitive SQUID magnetometry, there have been considerable theoretical and experimental studies carried out on the nanometer-scale magnets [1,2] which have been identified as candidates for the observation of macroscopic quantum phenomena (MQP) [3,4] such as the tunneling of the spin out of metastable potential minimum through the classically impenetrable barrier to a stable one, i.e., macroscopic quantum tunneling (MQT), or, more strikingly, macroscopic quantum coherence (MQC), where the spin coherently oscillates between energetically degenerate easy directions over many periods. In the semiclassical spin-coherent-state path-integral theory [5], MQC is connected with the presence of a topological term in the Euclidean action $S_E(\theta,\phi)$ arising from the nonorthogonality of spin coherent states, which is called Berry phase or the Wess-Zumino, Chern-Simons term: $iS\int_{-\frac{T}{2}}^{+\frac{T}{2}}(1-\cos\theta)\dot{\phi}(\tau)\,d\tau$, where S is the whole spin of the system, (θ,ϕ) are polar and azimuthal spin angles, repectively.

One of the manifestations of MQC is the ground-state tunnel splitting of magnetic systems. In the absence of an external magnetic field, it has been theoretically demonstrated that the ground-state tunnel splitting is completely suppressed to zero for the half-integer total spin ferromagnets or antiferromagnets with biaxial crystal symmetry [6,7], resulting from the destructive interference of the Berry phase in the Euclidean action between the symmetry-related tunneling paths connecting two classically degenerate minima. Such destructive interference effect for half-integer spins is known as the topological quenching. But for the integer spins, the quantum interference between topologically different tunneling paths is constructive, and therefore the ground-state tunnel splitting is nonzero.

While such spin-parity effects are sometimes related to Kramers degeneracy, they typically go beyond this theorem in rather unexpected ways. In the presence of an external magnetic field, as pointed out by Garg [8,9], the ground-state tunnel splitting can oscillate as a function of the field which is applied along its hard anisotropy axis in ferromagnets with biaxial crystal symmetry, and vanishes at certain values. This prediction is confirmed in one recent experiment carried out by Wernsdorfer and Sessoli [10]. They developed a new technique to measure the very small tunnel splitting on the order of 10^{-8}K in ferromagnetic molecular Fe_8 clusters. Indeed, they observed a clear oscillation of the tunnel splitting as a function of the magnetic field applied along the hard anisotropy axis, which is direct evidence of the role of the topological spin phase (Berry phase) in the spin dynamics of these molecules. Although this field induced oscillation's behaviour is investigated in great detail in ferromagnetic systems now [8–10], it is still less understood in antiferromagnetic molecular magnets such as Fe_{10}, Fe_6, V_8, and antiferromagnetic ferritin [11,12]. Golyshev and Popkov first studied MQC in a uniaxial antiferromagnetic fine particle in the presence of magnetic field [11], and found similar oscillation behavior. But they only calculated the Wentzel-Kramers-Brillouin (WKB) exponent in weak field approximation, and paid no attention to its preexponential factor. Later, in 1998, Chiolero and Loss considered the oscillation's properties of a ring-like molecular magnet using an anisotropic nonliner σ model (NLsM) [12]. In addition to the usual topological spin phase (Berry phase) term, they found a new quantum phase arising from fluctuations which is never seen in ferromagnetic molecular magnets. It is really a striking quantum property in antiferromagnetic molecular magnets. Unfortunately, the fundamental physics of this novel quantum phase is less explored so far.

In this paper, We would like to study the MQC of a biaxial symmetry antiferromagnetic molecular magnet

based on the two-sublattice model [13–15]. By applying the instanton technique in the spin-coherent-state path-integral representation [16], we obtain the rigorous instanton solutions and calculate both the WKB exponent and preexponential factor in the ground-state tunnel splitting. We will show that the quantum fluctuations around the classical paths can not only induce a new quantum phase previously reported by Chiolero and Loss [12], but also have great influence on the intensity of the ground-state tunnel splitting. Those features clearly have no analogue in the ferromagnetic molecular magnets. We suggest that they may be universal behaviors in all antiferromagnetic molecular magnets. Due to the instanton methods are semiclassical in nature, i.e., valid in large spins and in continuum limit, we perform exact diagonalization calculations and find that they agree well with the analytical results.

II. INSTANTON CALCULATIONS FOR SPIN-COHERENT-STATE PATH INTEGRALS

We consider the ring-like antiferromagnetic molecular magnets(i.e. Fe_{10}, Fe_6 and V_8) composed of $N = 2n$ spins s regularly spaced on a circle lying in the xy-plane with an antiferromagnetic exchange interaction between them [12,17]. In general, the crystalline anisotropy at each site has biaxial symmetry. As usually for antiferromagnets [13–15], we decompose the local spins into the two magnetic sublattices: $\vec{S}_1$ and $\vec{S}_2$ with the same spin value $S = ns$. Then, the molecular magnet in an external magnetic field $\vec{H}$ acting along its hard anisotropy axis can be described by a spin Hamiltonian of the type

$$\mathcal{H} = j\vec{S}_1 \cdot \vec{S}_2 + \left(k_1\hat{S}_{1z}^2 + k_2\hat{S}_{1y}^2 - g\mu_B H\hat{S}_{1z}\right) + \left(k_1\hat{S}_{2z}^2 + k_2\hat{S}_{2y}^2 - g\mu_B H\hat{S}_{2z}\right), \tag{1}$$

where g is the landé factor, and μ_B is the Bohr magneton. $k_1 > k_2 > 0$ are the crystalline anisotropy coefficients, and we take the easy, medium, and hard axes as **x**, **y**, and **z** respectively for each sublattice. j is the exchange energy. In accordance with experimental results it will be assumed that $j \gg k_1, k_2$ for the strong antiferromagnetic coupling. Note that our two-sublattice configuration is only valid for the magnetic field $H \leqslant H_a$. Here, $H_a = \frac{2jS}{g\mu_B}$ is the critical field at which the strong antiferromagnetic exchange interaction $j\vec{S}_1 \cdot \vec{S}_2$ is comparable to the Zeeman term $g\mu_B H(\hat{S}_{1z} + \hat{S}_{2z})$.

In the semiclassical approach [16], in order to obtain the ground-state tunnel splitting, one should compute the imaginary-time propagator in the spin-coherent-state representation:

$$\begin{aligned}&\langle \hat{n}_f \left| exp\left[-\mathcal{H}T\right] \right| \hat{n}_i \rangle \\ &= \int \mathcal{D}\Omega \exp\left(-S_E\right) \\ &= \int \mathcal{D}\{\theta_1\}\mathcal{D}\{\theta_2\}\mathcal{D}\{\phi_1\}\mathcal{D}\{\phi_2\} \exp\left(-\int d\tau \mathcal{L}\right)\end{aligned} \tag{2}$$

over all trajectories which connect the initial state $\mid \hat{n}_i\rangle$ to the final state $\mid \hat{n}_f\rangle$. Here θ_j, ϕ_j $(j = 1, 2)$ are the polar and azimuthal angles of each sublattice spin vector, the Lagrangian $\mathcal{L}$ include two parts [11]:

$$\mathcal{L}_0 = \sum_{j=1,2} iS(1 - \cos\theta_j)\dot{\phi}_j\,(\tau) \tag{3}$$

and

$$\begin{aligned}\mathcal{L}_1 = &J\left[1 + \cos\theta_1\cos\theta_2 + \sin\theta_1\sin\theta_2\cos\left(\phi_1 - \phi_2\right)\right] \\ &+ \sum_{j=1,2}\left(K_1\cos^2\theta_j + K_2\sin^2\theta_j\sin^2\phi_j\right) \\ &- \sum_{j=1,2} g\mu_B SH\cos\theta_j,\end{aligned} \tag{4}$$

corresponding to the Berry phase term and the total Euclidean energy term $E(\theta_1, \phi_1, \theta_2, \phi_2)$. Here, we have introduced $K_1 = k_1S^2$, $K_2 = k_2S^2$, and $J = jS^2$. All terms in (4) are of apparent physical meaning. The first term is the exchange interaction energy, the second term is magnetic anisotropy energy and the third term is Zeeman energy. The dominant contribution to the imaginary-time propagator comes from finite action solutions of the Euler-Lagrange equations of motion (instatons), which can be expressed as

$$\frac{\delta S_E}{\delta\bar{\theta}_j} = 0, \tag{5}$$

$$\frac{\delta S_E}{\delta\bar{\phi}_j} = 0, \tag{6}$$

where $\bar{\theta}_j$, $\bar{\phi}_j$ $(j = 1, 2)$ denote the classical paths.

According to the instanton technique in the spin-coherent-state path-integral representation [16], the instanton's contribution to the tunnel splitting Δ (not including the geometric phase factor generated by the Berry phase term in the Euclidean action) is given by

$$\Delta = p_0\omega_p\left(\frac{S_{cl}}{2\pi}\right)^{1/2} e^{-S_{cl}}, \tag{7}$$

where ω_p is the small-angle precession or oscillation frequency in the well, and S_{cl} is the classical action or the WKB exponent determined by Eqs. (5) and (6). The preexponential factor p_0 originates from the quantum fluctuations around the classical paths, which can be evaluated by expanding the Euclidean action to second order in the small fluctuations.

1. Wentzel-Kramers-Brillouin exponent

In our case, only low-energy trajectories with almost antiparallel $\vec{S}_1$ and $\vec{S}_2$ contribute the path integral. It is

therefore, safe to say that tunneling of $\vec{S}_2$ follows tunneling of $\vec{S}_1$ [14]. For that reason we can replace θ_2 and ϕ_2 by $\pi - \theta_1 + \varepsilon_\theta$ and $\pi + \phi_1 - \varepsilon_\phi$ respectively (with $|\varepsilon_\theta|, |\varepsilon_\phi| \ll 1$) in $\mathcal{L}$. In the new coordinates, the imaginary-time propagator of the system can be represented as

$$\int \mathcal{D}\{\varepsilon_\theta\}\mathcal{D}\{\varepsilon_\phi\} \int \mathcal{D}\{\theta_1\}\mathcal{D}\{\phi_1\} \times \exp\left(-\int d\tau \mathcal{L}\left(\theta_1, \phi_1, \varepsilon_\theta, \varepsilon_\phi\right)\right). \tag{8}$$

By simple algebra, up to the second order approximation about ε_θ and ε_ϕ, we obtain

$$\begin{aligned}\mathcal{L} = & \, i2S\dot{\phi} + 2\left(K_1 \cos^2\theta + K_2 \sin^2\theta \sin^2\phi\right) \\ & + \Big(-iS\dot{\phi}\sin\theta - K_1 \sin 2\theta \\ & + K_2 \sin 2\theta \sin^2\phi - g\mu_B SH \sin\theta \varepsilon_\theta\Big) \\ & + \left(K_2 \sin^2\theta \sin 2\phi\right)\varepsilon_\phi \\ & + iS\left[(1+\cos\theta) - \sin\theta\varepsilon_\theta\right]\dot{\varepsilon}_\phi \\ & + \left(A_{\theta\theta}\varepsilon_\theta^2 + A_{\theta\phi}\varepsilon_\theta\varepsilon_\phi + A_{\phi\phi}\varepsilon_\phi^2\right),\end{aligned} \tag{9}$$

where

$$\begin{aligned} A_{\theta\theta} &= -i\frac{S}{2}\cos\theta\dot{\phi} + \frac{J}{2} - K_1 \cos 2\theta \\ & \quad + K_2 \cos 2\theta \sin^2\phi - \frac{g\mu_B SH}{2}\cos\theta, \\ A_{\theta\phi} &= K_2 \sin 2\theta \sin 2\phi, \\ A_{\phi\phi} &= \frac{J}{2}\sin^2\theta + K_2 \sin^2\theta \cos 2\phi. \end{aligned} \tag{10}$$

In Eqs. (9) and (10), we have dropped the subscript of θ_1 and ϕ_1 for clarity. Upon Gaussian integrating (8) over ε_θ and ε_ϕ one can obtain the following effective Lagrangian

$$\begin{aligned}\mathcal{L}_{eff} = & \, i2S\dot{\phi} + 2\left(K_1\cos^2\theta + K_2 \sin^2\theta\sin^2\phi\right) + \\ & \frac{S^2}{2J}\left[\sin^2\theta\left(\dot{\phi} - ig\mu_B H\right)^2 + \dot{\theta}^2\right].\end{aligned} \tag{11}$$

Note that magnetic field enters only through the last term in Eq. (11) and has no influence on the tunneling barrier. Because of the condition $K_1 > K_2$, the equilibrium orientations of $\vec{S}_1$ are $(\theta, \phi) = \left(\frac{\pi}{2}, 0\right)$ and $\left(\frac{\pi}{2}, \pi\right)$ which correspond to two degenerate classical minima of the energy, $E = 0$. It is obvious from symmetry that there are two different type instanton trajectories of opposite windings around hard anisotropy axis. We denote them as $\pm$ instantons:

$$\phi = 0 \longrightarrow \phi = \pm\pi/2 \longrightarrow \phi = \pm n\pi \,(n = 1, 3, 5, ...). \tag{12}$$

To execute the first, we should seek the classical path(or paths) $\Omega_{cl}(\tau) = \left(\bar{\theta}(\tau), \bar{\phi}(\tau)\right)$ connecting the two minima, that minimizes the action $S_E = \int d\tau \mathcal{L}_{eff}$. This path satisfies the Euler-Lagrange equations of motion (see also Eqs. (5) and (6))

$$\frac{d}{d\tau}\left(\frac{\partial \mathcal{L}_{eff}}{\partial \dot{\Omega}_{cl}(\tau)}\right) - \frac{\partial \mathcal{L}_{eff}}{\partial \Omega_{cl}(\tau)} = 0. \tag{13}$$

Substituting the effective Lagrangian into Eq. (13), we obtain

$$\begin{aligned} & \frac{d}{d\tau}\left[\frac{S^2}{J}\frac{d\bar{\theta}}{d\tau}\right] \\ & = \left(-2K_1 + 2K_2 \sin^2\bar{\phi}\right)\sin 2\bar{\theta} \\ & \quad + \frac{S^2}{2J}\sin 2\bar{\theta}\left(\frac{d\bar{\phi}}{d\tau} - ig\mu_B H\right)^2 \end{aligned} \tag{14}$$

$$\begin{aligned} & \frac{d}{d\tau}\left[\frac{S^2}{J}\sin^2\bar{\theta}\left(\frac{d\bar{\phi}}{d\tau} - ig\mu_B H\right)\right] \\ & = 2K_2 \sin^2\bar{\theta}\sin 2\bar{\phi}. \end{aligned} \tag{15}$$

Consequently, a quasiclassical tunneling of $\vec{S}_1$ may occur in xy-plane $\bar{\theta} = \frac{\pi}{2}$, and then, Eq. (15) reduces to sine-Gordon equation,

$$2\frac{d^2}{d\tau^2}\bar{\phi} = \omega_1^2 \sin 2\bar{\phi}, \tag{16}$$

or equivalently

$$\frac{d\bar{\phi}}{d\tau} = \omega_1 \sin\bar{\phi}, \tag{17}$$

where $\omega_1 = \left(\frac{4JK_2}{S^2}\right)^{1/2}$. Under the boundary conditions in which the classical path approach the two minima as $\tau \to \pm\infty$, we obtain an exact solution of this equation [13],

$$\bar{\phi}(\tau) = 2\arctan\left(\exp\left(\omega_1 \tau\right)\right). \tag{18}$$

It is easily verified that $\bar{\phi} \to 0, \pi$, as $\tau \to \pm\infty$. The corresponding classical action, i.e., the WKB exponent in the rate of quantum tunneling at finite magnetic field, can be evaluated by integrating the Euclidean action with above classical trajectories, and the result is found to be

$$S_{cl}^{\pm} = \operatorname{Re} S_{cl} \pm i \operatorname{Im} S_{cl}, \tag{19}$$

with

$$\operatorname{Re} S_{cl} = 4S\left(\frac{K_2}{J}\right)^{1/2}, \tag{20}$$

$$\operatorname{Im} S_{cl} = 2\pi S\left(1 - \frac{H}{H_a}\right), \tag{21}$$

where the positive and negative sign in Eq. (19) are corresponding to $\pm$ instantons, respectively.

It is clearly seen from Eqs. (20) and (21), the classical action has two unusual features in the presence of magnetic field. First of all, the real part of action has no dependence on the magnetic field and is determined by material parameters of the system only. This feature is quite different from that in ferromagnetic molecular magnets, and can be understood easily from Eq.

(11), since the tunneling barrier remains unchanged under the magnetic field. Further, as shown from Eq. (21), if we ignore the contribution from quantum fluctuations around the classical paths, the ground-state tunnel splitting which is proportional to $\exp(-\operatorname{Re} S_{cl})\,|\cos(\operatorname{Im} S_{cl})|$ oscillates as the field H is increased, and the tunneling is thus quenched whenever

$$H = \frac{(2S-n-1/2)}{2S} H_a, \tag{22}$$

where $n = 0, 1, 2, \ldots$. It is interesting to note that this result agrees well with Eq. (11) in Ref. [8] found by Garg for ferromagnetic molecular Fe_8 clusters if one makes the replacement $J = 2S$ and sets $\lambda = 0$.

2. preexponential factor

The second major step is to evaluate the preexponential factor of small fluctuations around the classical instanton paths. We write

$$\theta(\tau) = \bar{\theta}(\tau) + \delta\theta(\tau), \tag{23}$$
$$\phi(\tau) = \bar{\phi}(\tau) + \delta\phi(\tau), \tag{24}$$

and evaluate the action to second order in $(\delta\theta, \delta\phi)$. Writing $S_E = S_{cl} + \delta^2 S$, we have

$$\begin{aligned}\delta^2 S = \int d\tau \frac{S^2}{2J} \Big\{ \delta\dot{\theta}^2 + \Big[(g\mu_B S H)^2 + \omega_0^2 - \omega_1^2 \\ + \omega_1^2 \cos 2\bar{\phi} \pm i2g\mu_B S H \omega_1 \sin\bar{\phi} \Big] \delta\theta^2 \\ + \delta\dot{\phi}^2 + \left(\omega_1^2 \cos 2\bar{\phi}\right) \delta\phi^2 \Big\},\end{aligned} \tag{25}$$

where $\omega_0 = \left(\frac{4JK_1}{S^2}\right)^{1/2}$. Note that the θ and ϕ fluctuations are decoupled in Eq (25), and in the θ-fluctuation, an unusual term $\pm i2g\mu_B S H\omega_1 \sin\bar{\phi}$ distinguishing + instantons from − instantons appears. As we will show below, this extra term has important consequences at the high magnetic field.

Now, the imaginary-time propagator is found to be

$$\langle \hat{n}_f \left| exp\left[-\mathcal{H}T\right] \right| \hat{n}_i \rangle = \exp\left(S_{cl}^{\pm}\right) D_{\delta\theta}^{\pm} D_{\delta\phi} \tag{26}$$

with

$$\begin{aligned} D_{\delta\theta}^{\pm} &= \mathcal{N}_\theta \int \mathcal{D}\{\delta\theta\} \times \\ &\exp\Big\{ -\int d\tau \Big[\frac{S^2}{2J}\delta\dot{\theta}^2 + \Big((g\mu_B S H)^2 + \omega_0^2 \\ &-\omega_1^2 + \omega_1^2 \cos 2\bar{\phi} \pm i2g\mu_B S H\omega_1 \sin\bar{\phi} \Big) \delta\theta^2 \Big] \Big\}, \end{aligned} \tag{27}$$

$$\begin{aligned} D_{\delta\phi} &= \mathcal{N}_\phi \int \mathcal{D}\{\delta\phi\} \times \\ &\exp\left\{ -\int d\tau \frac{S^2}{2J} \left[\delta\dot{\phi}^2 + \left(\omega_1^2 \cos 2\bar{\phi}\right) \delta\phi^2 \right] \right\}, \end{aligned} \tag{28}$$

where $\mathcal{N}_\theta$ and $\mathcal{N}_\phi$ are the normalization factors. The fluctuation determinant for ϕ is standard. Following the Eq. (2.44) in Ref. [16], we obtain

$$D_{\delta\phi} = 2\omega_1 \left(\frac{S^2 \omega_1}{J\pi} \right)^{1/2} = 2\omega_1 \left(\frac{\operatorname{Re} S_{cl}}{2\pi} \right)^{1/2}. \tag{29}$$

For the θ-fluctuation determinant we find in the first order perturbation theory (The detailed calculation of $D_{\delta\theta}^{\pm}$ will be reported elsewhere.), for the high magnetic field,

$$D_{\delta\theta}^{\pm} = \exp\left(\frac{\eta^{1/2} \mp i\frac{\pi}{2}}{\left(1+\frac{K_1}{K_2}\eta\right)^{1/2}} \right), \tag{30}$$

where $\eta = \frac{\omega_1}{g\mu_B S H} = \frac{H_a}{H}\left(\frac{K_2}{J}\right)^{1/2}$ is used as a small parameter in the high magnetic field. It is clearly shown in Eq. (30), the existence of magnetic field can bring a phase shift which approaches approximately $\frac{\pi}{2}$ in the high-field regime. Note that the value of phase shift agrees well with that found by Chiolero and Loss [12]. On the other hand, as the field decreases down to zero, and thus $\eta \to \infty$, the phase shift vanishes despite the breakdown of our first order perturbation calculations in the low-field regime.

Combing the classical action and two fluctuation determinants, we arrive at the desired ground-state tunnel splitting,

$$\begin{aligned}\Delta = 4\exp\left(\frac{\eta^{1/2}}{\left(1+\frac{K_1}{K_2}\eta\right)^{1/2}} \right) \omega_1 \left(\frac{\operatorname{Re} S_{cl}}{2\pi} \right)^{1/2} \times \\ \exp\left(-\operatorname{Re} S_{cl}\right) \left|\cos\Phi(H)\right|,\end{aligned} \tag{31}$$

where

$$\Phi(H) = 2\pi S \left(1 - \frac{H}{H_a}\right) + \frac{\frac{\pi}{2}}{\left(1+\frac{K_1}{K_2}\eta\right)^{1/2}}. \tag{32}$$

III. RESULTS AND DISCUSSIONS

The semiclassical analysis presented so far applies strictly speaking only to a sizable number of spins with $S \gg 1$. However, as is often the case with such methods the results are valid (at least qualitatively) even down to a few spins of small size. This expectation is indeed confirmed by exact diagonalization calculations which we have performed on Hamiltonian (1). Result for $S = 5$, and for some typical values $k_1 = 0.03$ K, $k_2 = 0.01$ K and $j = 1.0$ K is presented in Fig. 1, the critical field is found to be 7.44 T. Here, the units for the energy and magnetic field are taken to be Kelvin and Tesla, respectively. We can see that the numerical and semiclassical

approach show reasonable agreement in the whole magnetic field regime. Since our perturbed calculation for the θ-fluctuation determinant is only valid in the high magnetic field, the agreement in the low-field regime is surprising in some ways.

As shown in Fig.1, the ground-state tunnel splitting vanishs at the field $\frac{H}{H_a}=1.0$. This disappearance is evident for the extra $\frac{\pi}{2}$ phase shift, since according to Eq. (21), there should be a peak in usual. It is worthy noting that the extra $\frac{\pi}{2}$ phase shift is not limited to our biaxial symmetry antiferromagnetic molecular magnets case. Indeed, for the strong antiferromagnetic coupling, upon Gaussian integrating (8) over the small displacements ε_θ and ε_ϕ, one can obtain the effective Lagrangian in general form

$$\mathcal{L}_{eff} = i2S\dot{\phi} \\ +E\left(\theta,\phi\right)+\frac{S^2}{2J}\left[\sin^2\theta\left(\dot{\phi}-ig\mu_B H\right)^2+\dot{\theta}^2\right], \quad (33)$$

where the detailed form of $E\left(\theta,\phi\right)$ depends on the system investigated. Then, an extra term similar to $\pm i2g\mu_B SH\omega_1\sin\bar{\phi}$ in Eq. (25) will appear after we expand the small fluctuations around the classical instanton paths to the second order, and gives the $\frac{\pi}{2}$ phase shift. Thus, we conclude that the $\frac{\pi}{2}$ phase shift induced by θ-fluctuation may be a universal quantum behavior in all antiferromagnetic molecular magnets systems.

In the figure, another interesting feature is the peak height of the ground-state tunnel splitting. As the magnetic field increases, the peak height first drops significantly in the low-field regime, and then keeps invariant up to the critical field H_a. This may be qualitatively understood from Eq. (21). Because the WKB exponent has no relevance with the magnetic field, the most essential dependence of the ground-state tunnel splitting on the field comes from the preexponential factor $p_0=4\exp\left(\frac{\eta^{1/2}}{\left(1+\frac{K_1}{K_2}\eta\right)^{1/2}}\right)$ which undergoes a dramatically change only in the low-field regime.

At the end of this section, in order to support the experimental relevance of our results, we give some estimates for the ferric wheel, Fe_{10}, for which $N=2n=10$, $s=\frac{5}{2}$, $S=ns=12.5$. If one takes $\frac{j}{g\mu_B}=4$ T, $\frac{k_1}{j}=0.03$ and $\frac{k_2}{j}=0.01$ as the typical parameters values [12], then the simple algebra demonstrates that the ground-state tunnel splitting has $2S=25$ oscillations of magnitude $\Delta\approx 1.3$ K and period 4 T. From Eq. (30), the phase shift will be visible when $\frac{K_1}{K_2}\eta\sim 3$, or $H=10$ T. Therefore, all quantities appear to be well within experimental reach.

IV. CONCLUSION

In summary, we have investigated the MQC phenomena in biaxial symmetry antiferromagnetic molecular magnets. Our discussion is based on the two-sublattice model that includes anisotropy and magnetic field. On the basis of instanton technique in the spin-coherent-state path-integral representation, both the rigorous Wentzel-Kramers-Brillouin exponent and preexponential factor for the ground-state tunnel splitting are obtained. We have outlined here two prominent features in our tunneling scenario: (i) In addition to the usual topological term in the classical action, a new quantum phase arising from the quantum fluctuations around the classical paths is found to contribute to the tunneling oscillations. This result coincides with that reported by Chiolero and Loss [12]. (ii) The magnetic field appears to have no influence on the tunneling barrier. Thus the main dependence of the tunneling peak height on the field comes from the quantum fluctuations, this leads to a sudden drop of peak's height in the low-field regime. Both two features clearly have no analogue in the ferromagnetic systems [8–10]. We suggest that they may be universal behaviors in all antiferromagnetic molecular magnets.

We realize that our result is based on the instanton method which is semiclassical in nature, i.e., valid in large spins and in continuum limit. We perform exact diagonalization calculation to check its validity, and find that it agrees well with the analytical result in the regime where a comparison is possible.

Recent experiments have rekindled interest in the field of quantum tunneling of the molecular magnets.. Most notable has been the discovery of resonant quantum tunneling between spin states in the ferromagnetic system of spin-10 molecules such as $Mn_{12}Ac$ [1] and Fe_8 [2]. Since the antiferromagnetic molecular magnets are proposed as better candidates for observing the phenomena of MQT and MQC [13] compared with the ferromagnetic ones, we hope that our predictions on antiferromagnetic molecular magnets can be confirmed in experiments in the future.

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Figure caption

Fig.1. $\frac{\Delta}{J}$ versus $\frac{H}{H_a}$, with $S = 5$, $k_1 = 0.03$ K, $k_2 = 0.01$ K, $j = 1.0$ K and $H_a = 7.44$ T. Here, the units for the energy and magnetic field are taken to be Kelvin and Tesla. The solid line and symbols represent, respectively, the ground-state tunnel splitting predicted by the semiclassical instantons approach and that obtained by the exac diagonalization method.

Fig.1
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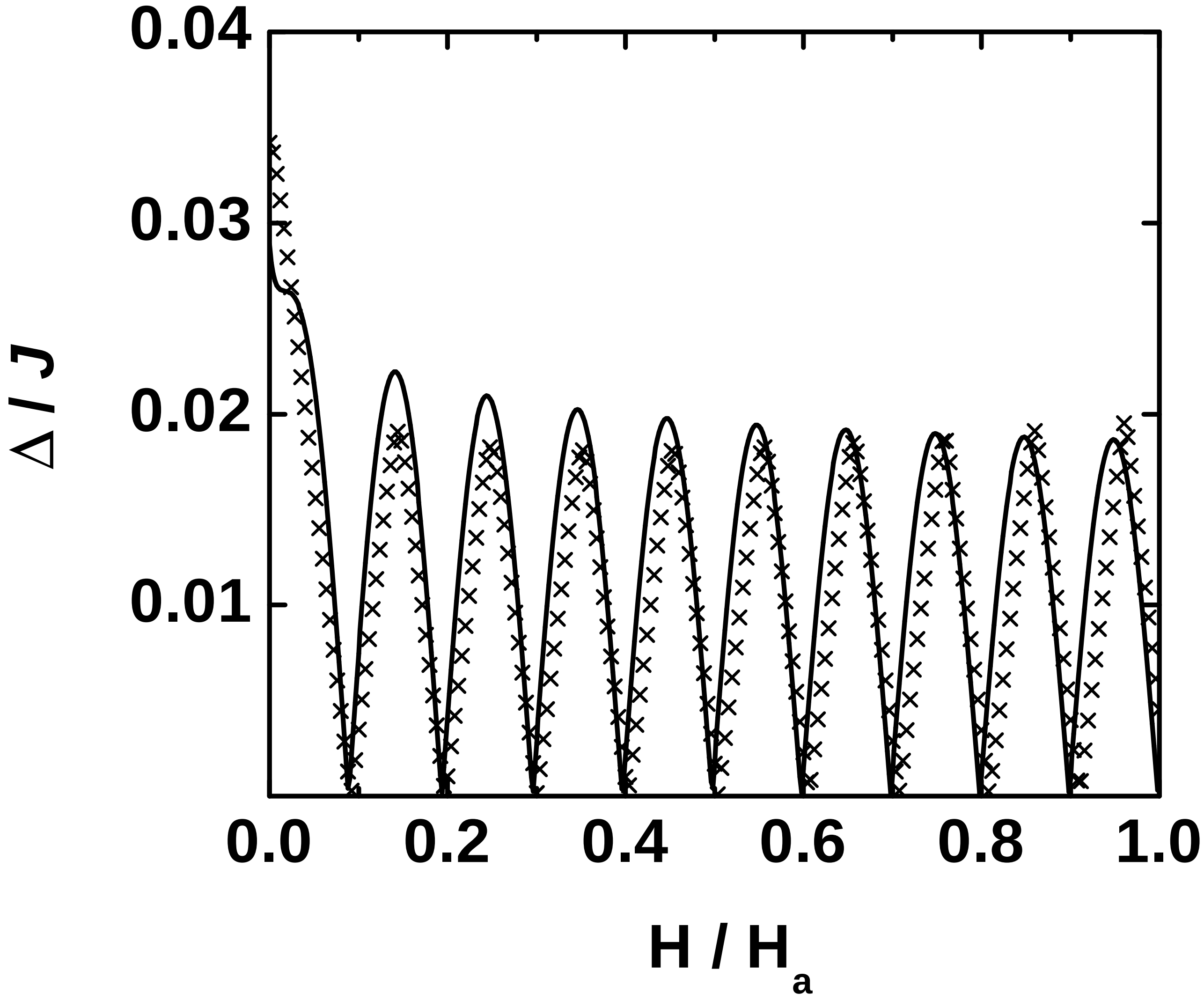

Fig.1